Combustion Research Facility News

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Broadly tunable, sensitive infrared spectrometer developed

Infrared absorption spectroscopy is a powerful tool for in-situ sensing of gases, particularly when using high-brightness laser sources. One issue that has hindered the broad use of this technique is the availability of well-behaved IR lasers. Diode lasers are useful, but tune only over narrow spectral regions. Pulsed, nonlinear methods produce broader tuning at the expense of size and complexity. Recently, Ken Aniolek, Peter Powers (University of Dayton), Tom Kulp, Bruce Richman, and Scott Bisson have merged three new technologies to produce an infrared spectroscopic system that alleviates these problems.

The new approach combines quasi-phasematching (QPM) and cavity-ringdown spectroscopy (CRS). QPM is a method to "engineer" nonlinear crystals to phase-match at prescribed wavelengths and at the nonlinearity of the material. This yields crystals having very broad tunability and high gain. These attributes were exploited to generate the light source shown in Fig. 1. It consists of an optical parametric generator (OPG), an electronically-tunable spectral filter, and an optical parametric amplifier (OPA). The QPM crystal is PPLN. The OPG generates a broad signal and idler emission in a single pass. This is filtered and then used to seed the single-pass OPA, yielding spectrally-narrow (0.05 cm^{-1}) signal and idler beams. The filter bandpass can be rapidly slewed over the PPLN bandwidth, which is determined by the crystal periodicity. By combining electronic tuning and translation of different periodicities through the beam, seamless tuning over several hundred cm^{-1} is possible, selectable anywhere between 2200 and 7700 cm^{-1}. Most importantly, because the system involves no laser cavities, it eliminates the need for active mode stabilization.

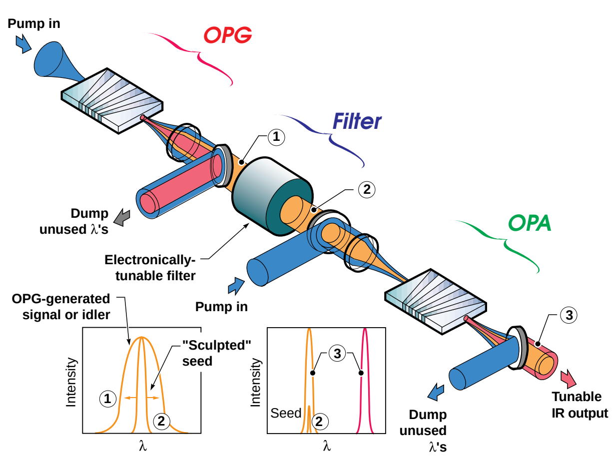

Figure 1. Diagram of the OPG/OPA apparatus.

Due to its self-normalizing nature, CRS is ideally suited for high-sensitivity absorption measurements using pulsed lasers. Conversely, the new light source is ideally suited to CRS. Because a single "ringdown" lasts ~10 μs, CRS is optimally performed at high pulse repetition rates. The low PPLN OPG threshold makes it possible to efficiently pump the OPG/OPA with a high repetition rate diode-pumped Nd:YAG laser. Currently, the use of a 1 kHz rep-rate laser allows single-line scanning in ~50 ms and a 15-cm^{-1} region in ~1 s. Figure 2 shows a 70-cm^{-1} "mosaic" scan of a 500-ppb methane sample.

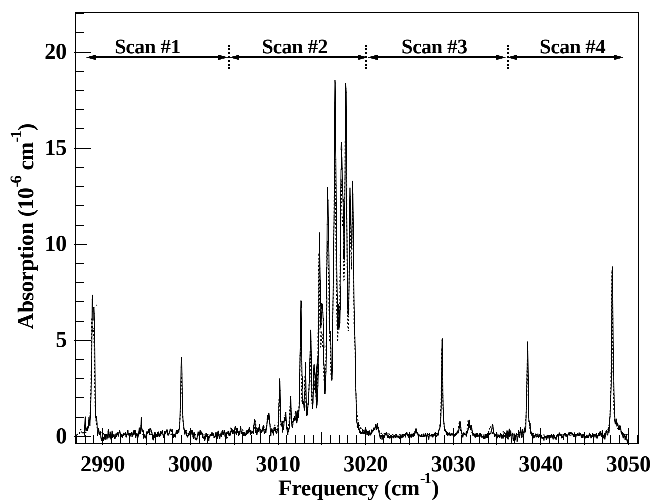

Figure 2. Spectral measurement of 500-ppb methane collected with the ringdown apparatus. The measured spectrum is overlapped with a calculated spectrum (dashed line). Fine wavelength scanning was accomplished by tuning the electronic filter; coarse scanning was accomplished by translating the beams to four different regions (periodicities) of the PPLN crystals.

CRB now on web

The Combustion Research Bulletin was formerly distributed via the mail. It is now available on the CRF website, http://www.ca.sandia.gov/CRF/Publications/CRB/CRB_4.html

Jay Keller (left) and Dennis Witmer, Univ. of Alaska, Fairbanks look over a 3kW proton exchange membrane fuel cell (PEMFC). This PEMFC is part of a new collaborative program designed to develop and deploy domestic energy systems for application in remote arctic Alaskan villages. Using an integrated energy and materials management systems approach, a total fuel energy utilization of 80 percent is expected. Additional benefits include: supplying potable water, reduced emissions, and system reliability.

Michael Försth (center), a doctoral student at Chalmers Univ. of Technology, Sweden, has completed experiments examining charge mixing in spark ignition engines with Pete Hinze (left) and Paul Miles (right). Michael's work involved in-cylinder measurements of charge temperature and composition using spontaneous Raman scattering.

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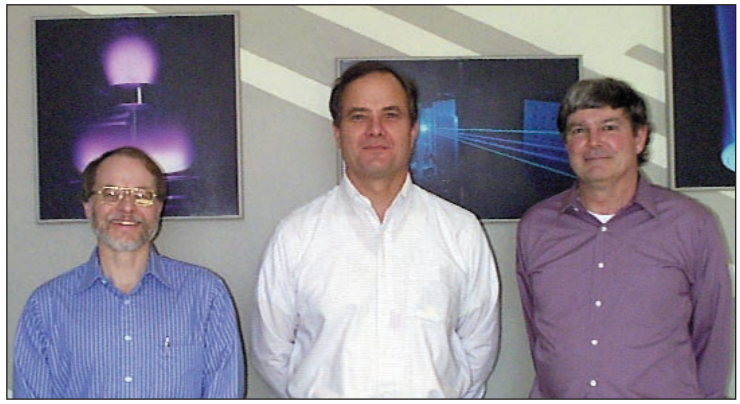
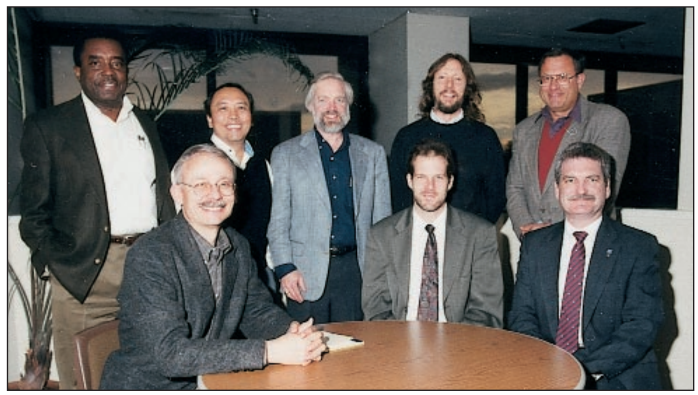


John Dec (left), Dennis Siebers (center), and Pete Witze were recently elected to the Fellow grade of membership in the Society of Automotive Engineers. More information regarding this prestigious recognition and each individual can be found on the CRF website, www.ca.sandia.gov/crf/WhatsHot/Whatshot.html.



The inaugural meeting of the CRF User Executive Committee was held on November 24, 1998. The committee includes (seated l to r): Prof. Phil Smith, Univ. of Utah; Prof. Bob Hurt, Brown Univ.; Roy Primus, Cummins Engine Co. and standing (l to r): Vince Henry, Visteon/Ford Automotive Glass; Prof. J. Y. Chen, Univ. of California at Berkeley; Prof. Houston, Cornell Univ.; Prof. Marshall Long, Yale Univ.; and Charlie Westbrook, Lawrence Livermore National Laboratory. Prof. Jim Driscoll, Univ. of Michigan, was unable to attend.

Prof. Terry Cool (left), Cornell Univ., recently assisted Andy McIlroy in developing a new low-pressure flame molecular beam spectrometer for the study of flame chemistry. The new instrument will be used first to study polyaromatic hydrocarbon formation in rich flames.

Hydrodynamic Instability in Liquid Propellants Modeled

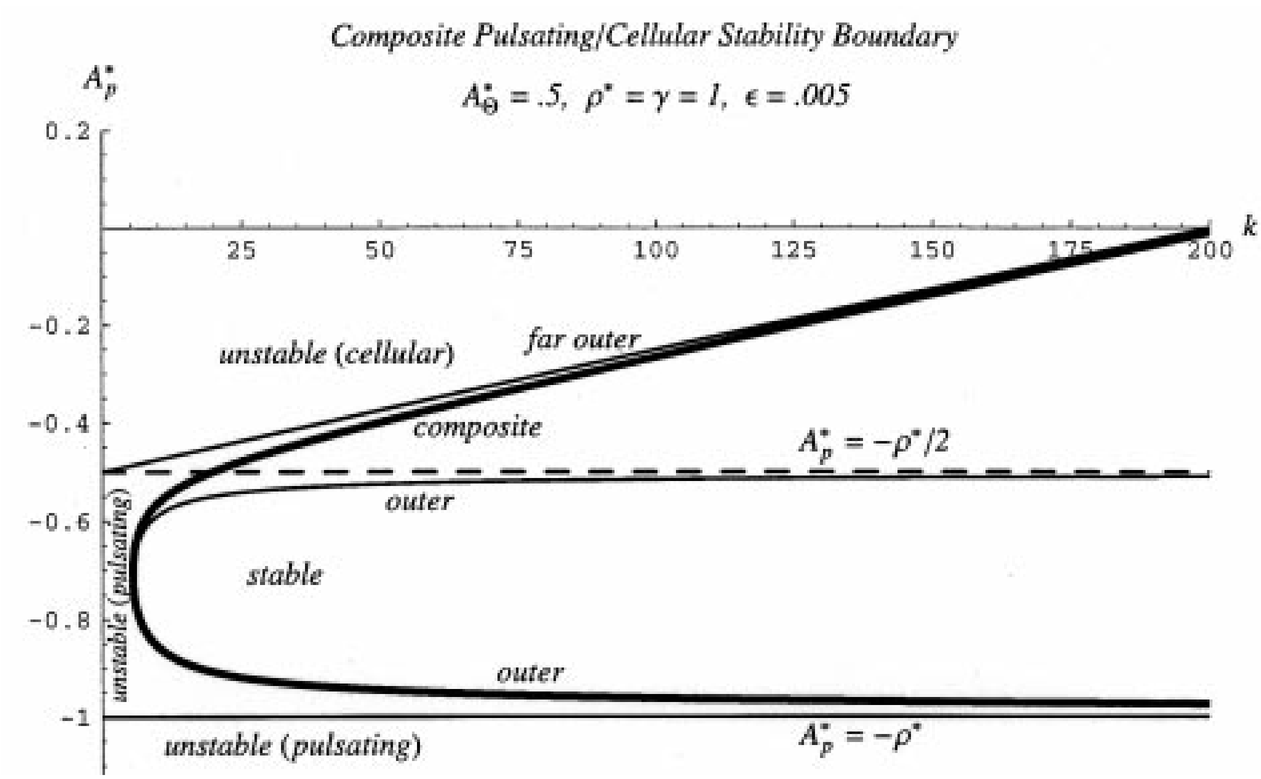
Steve Margolis, with support from NASA's Microgravity Science Research Program, has significantly extended the models for liquid propellant combustion to account for, among other things, local variations in the burning rate arising from pressure and temperature perturbations at the liquid/gas interface. Analyses of these extended models have led to considerable refinements in our understanding of hydrodynamic instability in liquid-propellant combustion under both normal and reduced gravity conditions.

The linear stability analysis of steady, planar burning, and the subsequent derivation of the dispersion relation that determines the neutral stability boundaries, were greatly facilitated by introducing appropriate scalings based on the smallness of the gas-to-liquid density ratio. As a result, there emerge three distinguished disturbance-wavenumber regimes (for small, intermediate and large wavenumbers k) in which one or more effects assume dominance over the others. In particular, for the classical cellular (Landau) form of hydrodynamic instability, gravity is a significant stabilizing effect for small-wavenumber disturbances, while surface tension and viscosity are the dominant stabilizing influences in the large-wavenumber region. Results for both the special inviscid limit as well as the fully viscous case indicate the existence of a critical value of the pressure sensitivity A_p for the onset of cellular hydrodynamic instability.

In the microgravity limit, the location of the critical value of $A_p(k)$ corresponding to the onset of instability shifts from order unity values of k to small disturbance wavenumbers (*i.e.*, large disturbance wavelengths). Thus, the essential difference between Landau instability at normal and reduced gravity is that in the latter case, it becomes a long-wave hydrodynamic instability. In that case, the break-up of the liquid/gas interface is likely to occur via the formation of large cells on the liquid surface.

In addition to the classical cellular instability, which was the type of instability that emerged in the original Landau/Levich models, a pulsating form of hydrodynamic instability has been shown to exist for sufficiently negative values of the pressure sensitivity. In the inviscid limit, this boundary is simply a straight line, as depicted in the Figure, but in the viscous case, it was shown that the effects of viscosity are stabilizing for all but small wavenumbers.

A more significant effect is felt when, in addition to the pressure sensitivity A_p, the temperature sensitivity A_Θ of the burning rate is also allowed to be nonzero. In that case, for sufficiently large values of the latter, the pulsating boundary develops a turning point as illustrated in the Figure. Under these conditions, the stable parameter regime is lost and steady, planar combustion becomes intrinsically unstable to small-wavenumber pulsating disturbances. Values of the ratio A_Θ / A_p over which the development of this turning point occurs turn out to be roughly of the order of the overall activation energy. Consequently, a pulsating form of hydrodynamic instability, rather than the classical cellular (Landau) type, may be the more likely manifestation of hydrodynamic instability in at least some types of liquid propellants.



Composite hydrodynamic stability boundary (heavy line) for a sufficiently large value of the temperature-sensitivity parameter A_Θ. The lighter curves, to which the composite boundary asymptotes, denote the inviscid pulsating and cellular boundaries for $A_\Theta = 0$ in the far outer wavenumber region.

Thermal Decomposition of HMX Investigated

Sabrina Mack, Jeanette Wood, and Richard Behrens are examining the physical and chemical processes involved in the solid-phase thermal decomposition of HMX. The simultaneous thermogravimetric modulated beam mass spectrometry technique is used to gather data on the identities and rates of formation of the decomposition products. Additionally, scanning electron microscopy (SEM) is used to examine the morphology of HMX particles at different points in the decomposition process. The data obtained from the experiments are used in the development of models for the response of HMX to abnormal thermal conditions, such as a fire.

The SEM shown in Figure 1 is an example of a partially decomposed HMX particle. It illustrates the morphology of a cleaved particle that has undergone 32% decomposition followed by sublimation of 85% of the remaining HMX to reveal the structure of the non-volatile residue. This residue, taking the form of shells, represents the remnants left from reaction processes taking place within bubbles in microscopic regions. The main factor in determining the growth of the bubbles is the pressure of the gaseous products contained within. As the reaction proceeds, the bubbles grow within the grains until a grain boundary is intercepted and the gases are released. The porosity resulting from decomposition may lead to convective burning after ignition, which may cause a more violent event.

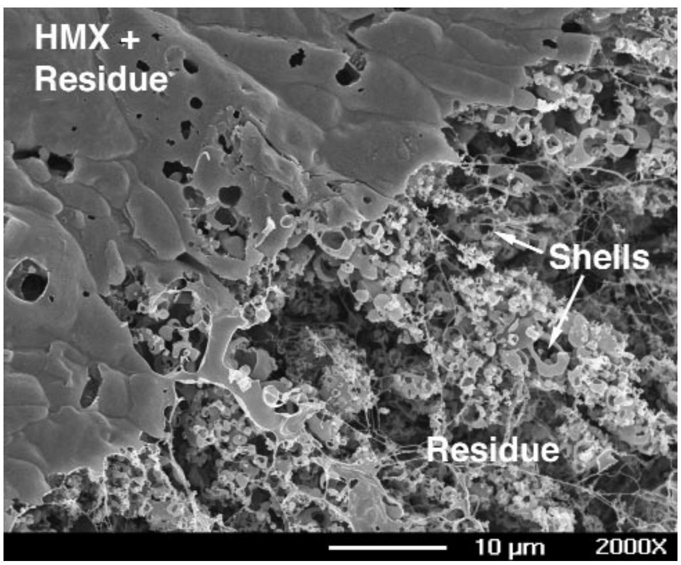


Figure 1. Detailed structure of the non-volatile residue of an HMX particle that has undergone 32% decomposition followed by sublimation of 85% of the remaining HMX.

Figure 2 shows the percentage of the total number of shells as a function of particle diameter for data gathered at two points in the decomposition process. The width of the grains appears to be correlated with the shell sizes for the 3% decomposed sample. This supports the idea that at this stage in the decomposition, the growth of the bubbles stops and the gaseous products are released. Transgranular bubble growth may account for the larger shell sizes plotted for the 32% decomposed sample.

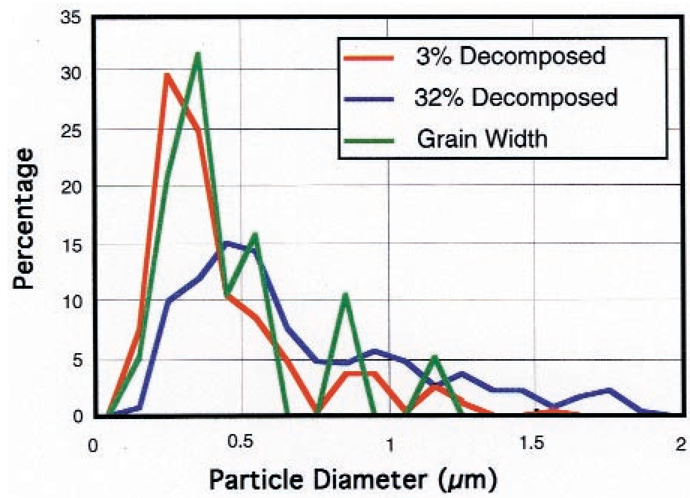


Figure 2. Shell and layer size distribution at two points during the decomposition of HMX.

The size, number density, and location of the shells have provided information on the reactive processes controlling the decomposition. The reactive processes determine the pre-ignition state of the material, therefore characterization of these processes is important to the development of the models. Future research on larger samples and HMX-based explosives and propellants will aid in the development of accurate models.

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